

## **Fundamentals of Rayleigh Scattering For Aerodynamic Measurements**

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### **I. Introduction**

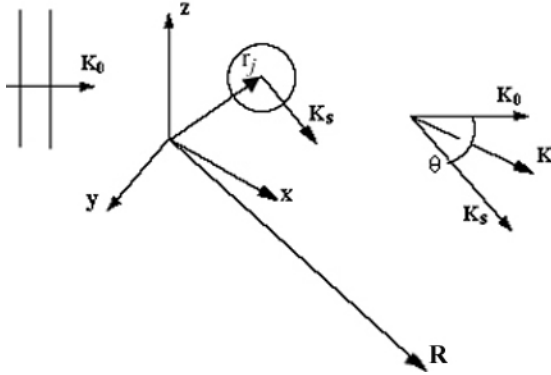
In the recent years Rayleigh scattering based techniques have found increasing applications for aerodynamic measurements<sup>1-5</sup>. A single measurement of Rayleigh spectra provides information of fluid density, temperature and one component of velocity. In addition, the absence of any intrusive probe and seed particles (used in the Mie scattering based techniques) makes Rayleigh scattering based techniques ideal non-intrusive diagnostic tools for a wide velocity (very low to hypersonic velocity) and temperature ranges. The present paper jots down the fundamental physical principles of the Rayleigh scattering process which was first treated by Lord Rayleigh and then by a host of other researchers.

When light waves are allowed to pass through a gas, the molecules present in the gas produce inelastic and elastic light scattering. The inelastic part is associated with a frequency shift from the incident light and is called Raman scattering, while the elastic part with no appreciable shift (except for the Doppler shift from moving gas molecules) is called Rayleigh scattering. This, however, is a loose separation as recent critical analyses includes the rotational part of the Raman scattered light in Rayleigh spectra<sup>6</sup>. If there exists coherent motion among gas molecules, such as that caused by very high frequency sound waves (induced by random thermal motion) and thermal diffusion, then additional scattering process, known as Brillouin scattering sets in. In fact even the Rayleigh scattering from an agglomeration of molecules will produce little scattering everywhere except in the forward direction without the presence of this coherent molecular motion. Therefore, in reality, when the elastic part of the scattered light is analyzed, one encounters both Rayleigh and Brillouin scatterings<sup>7, 8</sup>. In the following we will first analyze light scattered by single molecule and come up with the expression for the total scattered light. Subsequently, we will analyze the combined effect from an agglomeration of molecules and derive expressions for Rayleigh spectrum. The paper excludes analysis of Brillouin scattering, which hopefully will be analyzed in future.

### **II. Power of Rayleigh scattered light**

The scattering diagram from a single molecule is shown in Fig. 1. The first assumption is that the molecule is spherical, such as in the case of noble gases, Helium, Argon etc. Scattering from the commonly encountered diatomic molecules is somewhat more involved, although the general insights obtained from spherical molecules holds for the most part (see reference 6). Let's assume that the incident electric field is from plane parallel waves of wave number  $\mathbf{k}_0$  and frequency  $\omega_0$ .

$$E = E_0 \exp \left[ i \left( \mathbf{k}_0 \cdot \mathbf{r}_j - \omega_0 t \right) \right] \quad (1)$$



**Fig. 1** Diagram of scattering geometry, where  $\mathbf{r}_j$  is the location of a single scatterer,  $\mathbf{R}$  is the location of observer. The incident scattered and scattering vectors are represented by  $\mathbf{k}_0$ ,  $\mathbf{k}_s$  and  $\mathbf{k}$ , respectively.

The light wave produces an oscillating polarization  $P(\mathbf{r}, t)$  which radiates an electric field to the field point  $\mathbf{R}$  as

$$E_{ind} = \frac{\ddot{P}\left[\mathbf{r}_j, t - \frac{|\mathbf{R} - \mathbf{r}_j|}{c}\right] \sin \chi}{c^2 |\mathbf{R} - \mathbf{r}_j|} \quad (2)$$

where  $\sin \chi$  is the angle between the polarization vector of the incident wave and the propagation vector of the scattered wave  $\mathbf{k}_s$ . Now polarization of the molecule is dependent on the polarizability constant  $\alpha$ :  $P = \alpha E$ . Polarizability is truly a function of space and time for a dense gas but we will neglect this variation for the present paper. For  $r_j \ll R$  the scattered electric field is

$$E_{ind} = -E_0 \left(\frac{\omega_0}{c}\right)^2 \frac{\exp[i(\mathbf{k}_s \cdot \mathbf{R} - \omega_0 t)] \sin \chi}{R} \alpha \exp[i(\mathbf{k}_0 - \mathbf{k}_s) \cdot \mathbf{r}_j] \quad (3)$$

The power of the scattered light is following:

$$I_{ind} = \frac{\epsilon_0 c}{2} E_0^2 \left(\frac{\omega_0}{c}\right)^4 \frac{\alpha^2 \sin^2 \chi}{R^2} = I_0 \frac{16\pi^4}{\lambda^4} \frac{\alpha^2 \sin^2 \chi}{R^2} \quad (4)$$

where,  $I_0$  is the incident light intensity,  $\lambda$  is vacuum wavelength. This equation provides significant insight into the characteristics of the Rayleigh scattered light. First the scattered light intensity is inversely proportional to  $\lambda^4$ , i.e., the smaller the wavelengths the higher the scattered light intensity. This is the reason for blue color of sky and that of deep clear water. For aerodynamic measurements it is desirable to use ultra-violet laser rather than visible or infrared. UV lasers have been used by various researchers; however, such lasers have a limited power output and the available optical components are also limited. Therefore, most of the Rayleigh diagnostics use blue or green lines from gas and frequency-doubled solid state lasers.

The second important aspect of equation 4 is the polarization dependence via  $\sin^2 \chi$  term. To maximize scattered light collection, the collection optics should be placed in a plane  $90^\circ$  to the incident polarization of the laser beam. This is easily obtained by

rotating a half wave plate placed on the incident laser path. If optical fibers are used to deliver light, then polarization preserving fibers are preferred over multi-mode type, as the latter scrambles the incident laser polarization.

The polarizability constant  $\alpha$  is a property of the gas and varies from species to species. It is related to the refractive index  $n$  (or dielectric constant  $\epsilon = n^2$ ) via Lorentz-Lorenz Formula<sup>9</sup>:

$$\alpha = \frac{3}{4\pi N} \frac{\epsilon - 1}{\epsilon + 2} = \frac{3}{4\pi N} \frac{n^2 - 1}{n^2 + 2} \quad (5)$$

where,  $N$  is number density (number per unit volume) of molecules. For air  $n \sim 1$  and

$$\alpha = \frac{1}{2\pi} \frac{n-1}{N} \quad (6)$$

It should be noted here that  $(n-1)/N$  is independent of Number density as the refractive index  $n$  is also a function of Number density; therefore,  $\alpha$  is a molecular property.

Combining equations 4 and 6:

$$I_{ind} = I_0 \frac{4\pi^2}{\lambda^4} \frac{\sin^2 \chi}{R^2} \left( \frac{n-1}{N} \right)^2 \quad (7)$$

It is of interest to determine the total power of the scattered light produced by a single molecule. This is can be determined by integrating equation 7 at all polarization angles  $\chi$  over an enclosing sphere of radius  $R$ :

$$I_{tot} = I_0 \frac{32\pi^3}{3\lambda^4 R^2} \left( \frac{n-1}{N} \right)^2 \quad (8)$$

However, the commonly used parameter is Rayleigh scattering cross section  $\sigma$ , which is defined as a differential of solid angle along the collection direction:  $\partial\sigma/\partial\Omega$ .

$$I_{ind} = I_0 \frac{1}{R^2} \frac{\partial\sigma}{\partial\Omega} \quad (9)$$

$$or, \frac{\partial\sigma}{\partial\Omega} = \frac{4\pi^2}{\lambda^4} \left( \frac{n-1}{N} \right)^2 \sin^2 \chi$$

$\partial\sigma/\partial\Omega$  is a property of the individual gas or gas mixture and the wavelength of the incident light. The values of the Rayleigh scattering cross-section is tabulated in various articles. For aerodynamic measurements, properties of air is of interest. Some of the sample numbers are tabulated in Table 1. Note that the values provided in that table are in terms of  $\sigma$ . To follow the definition of equation 9 which is in terms of cross-section per unit of solid angle  $\Omega$ , one needs to divide the quoted numbers by  $4\pi$ .

**Table 1. Light wavelength and Rayleigh scattering cross-section of standard air (from reference 6)**

Wavelength (nanometer)	Rayleigh scattering Cross-section $\sigma$ (cm <sup>2</sup> )
200	$3.612 \times 10^{-25}$
300	$5.642 \times 10^{-26}$
400	$1.673 \times 10^{-26}$
500	$6.656 \times 10^{-27}$
530	$5.245 \times 10^{-27}$
600	$3.161 \times 10^{-27}$
800	$9.854 \times 10^{-28}$
1000	$4.010 \times 10^{-28}$

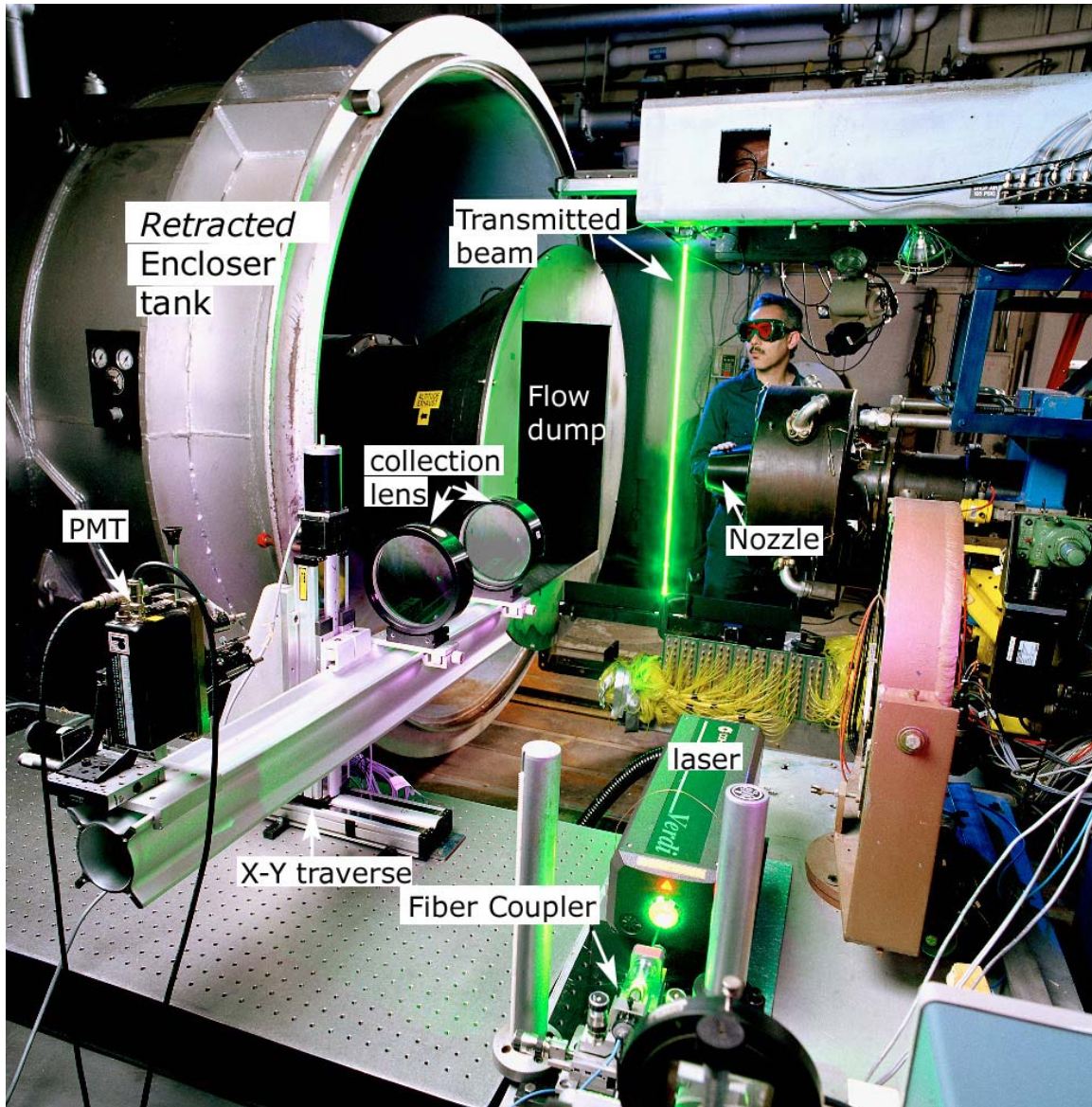
When Rayleigh scattering is used for diagnostics, (fig. 2), the scattered light is collected by a lens whose  $f/\#$  determines the collection solid angle  $\Delta\Omega$ . In that case power collected from a single scatterer is:

$$\Delta I = I_0 \int_{\Delta\Omega} \frac{\partial \sigma}{\partial \Omega} \partial \Omega \quad (10)$$

Now light is scattered over a volume  $\Delta v$  of the incident laser beam and if the molecular number density is  $N$  then the total number of scatterers are  $N \Delta v$ . For the purpose of determining the power of the collected light,  $I_{coll}$  it is sufficient to sum the contribution from individual molecules:

$$I_{coll} = \eta I_0 N \Delta v \int_{\Delta\Omega} \frac{\partial \sigma}{\partial \Omega} \partial \Omega \quad (11)$$

where,  $\eta$  is the efficiency of the collection system. It is the number density  $N$  which is related to local gas density. For a fixed optical system and gas composition, the collected light power becomes a function of density only. This has been the basic principle for measuring air density using Rayleigh scattering principle. On the other hand, if the density is nearly constant but Rayleigh cross-section is changed due to different concentration of say Methane or Helium gas in the air, then the collected power is related to the mole fraction of these additives. This principle has been used to study turbulence mixing between two streams or combustible mixtures<sup>1</sup>.



**Fig. 2** A Rayleigh scattering setup in a Nozzle testing Facility of NASA Glenn Research center (reference 5). About 0.5 Watt of laser light from a frequency doubled Nd:VO<sub>4</sub> laser (532nm) was passed into the large tank (now retracted back to the left) enclosing the Nozzle facility via polarization preserving fiber. Light scattered from various points along the beam was collected and measured using photo-multiplier tube to determine gas density variations across the supersonic plume.

### III. Spectral distribution of Rayleigh Scattered Light from a gas flow:

When light is scattered by a large number of molecules, there appears two additional issues. First, the phase difference of the scattered light from various molecules needs to be considered. Second, Doppler shift from the random thermal motion and the bulk motion of the gas flow needs to be considered. Both of these distribute the scattered light in frequencies other than the incident laser frequency and gives rise to a spectral distribution. The first issue gives rise to Brillouin scattering, which will not be addressed in this paper. The second issue is used to measure the temperature and bulk motion of the

gas flow which is addressed in the following. It should be pointed out that the frequency changes are far less than that seen from the vibrational Raman scattering, but may overlap some of the rotational Raman Lines<sup>6</sup>. The starting point for spectral calculation is equation 3, where electric field from a single molecule is described as a function of the polarizability constant. To derive Rayleigh spectra from dilute gases, we will once again assume a constant  $\sigma$ . Equation 3 is rewritten as:

$$E_{ind} = E_i \exp[i(\mathbf{k}_s \cdot \mathbf{R} - \omega_0 t)] \exp[i(\mathbf{k} \cdot \mathbf{r}_j)],$$

$$\text{where, } \mathbf{k} = \mathbf{k}_0 - \mathbf{k}_s, \quad \text{and } E_i = -\frac{E_0 \alpha}{R} \left( \frac{\omega_0}{c} \right)^2 \quad (12)$$

$\mathbf{k}$  is the scattering vector as shown in fig. 1. Since, the wave-number of the scattered light is nearly the same as that of incident light

$$|\mathbf{k}| = 2|\mathbf{k}_0| \sin \frac{\theta}{2} \quad (13)$$

To construct spectra the first step is to determine an auto-correlation function  $R_E(\tau)$ , where  $\tau$  is the delay time.

$$R_E(\tau) = \lim_{T \rightarrow \infty} \frac{1}{2T} \int_{-\infty}^{\infty} E_{ind}(R, t + \tau) \cdot E_{ind}^*(R, t) dt$$

$$= E_i^2 \lim_{T \rightarrow \infty} \frac{1}{2T} \int_{-\infty}^{\infty} \exp[-i\omega_0 \tau] \exp[i\mathbf{k} \cdot (\mathbf{r}_j(0) - \mathbf{r}_j(\tau))] dt \quad (13)$$

The normalized (by the mean square of correlation i.e.,  $R_E(0)$ ) spectral density is calculated via a Fourier transform:

$$S(\mathbf{k}, \omega) = \frac{\int_{-\infty}^{\infty} R_E(\tau) \exp(i\omega \tau) d\tau}{R_E(0)} = \int_{-\infty}^{\infty} d\tau \exp[i(\omega - \omega_0)\tau] \int_{-\infty}^{\infty} \exp[i\mathbf{k} \cdot (\mathbf{r}_j(0) - \mathbf{r}_j(\tau))] dt \quad (14)$$

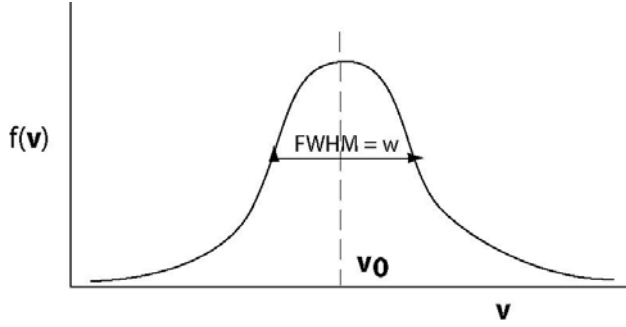
The next step is to determine the spectral shape due to moving gas molecules with a velocity  $\mathbf{v}$ . For a moving molecule with speed  $\mathbf{v}$ , the term  $\mathbf{k} \cdot \mathbf{r}_j(\tau) = \mathbf{k} \cdot \mathbf{r}_j(0) + \mathbf{k} \cdot \mathbf{v} \tau$ .

$$S(\mathbf{k}, \omega) = \int_{-\infty}^{\infty} d\tau \exp[i(\omega - \omega_0 + \mathbf{k} \cdot \mathbf{v})\tau] \int_{-\infty}^{\infty} \exp[i\mathbf{k} \cdot (\mathbf{r}_j(0) - \mathbf{r}_j(0))] dt \quad (15)$$

The second exponential term represents phase difference from two molecules at  $\mathbf{r}_j$  and  $\mathbf{r}_i$  and leads to Brillouin scattering. For the subsequent discussion, only the first integral leading to Rayleigh spectra from velocity distribution alone will be considered. In a gas the molecules move over a wide range of speed. In addition for a moving gas there is a superimpose bulk velocity  $\mathbf{v}_0$ . For a dilute gas (away from critical point) the velocity distribution is Maxwellian<sup>10</sup> (Fig. 3)

$$f(\mathbf{v}) d^3 \mathbf{v} = \frac{1}{\pi^{\frac{3}{2}} w^3} \exp \left[ -\frac{|\mathbf{v} - \mathbf{v}_0|^2}{w^2} \right] d^3 v \quad (16)$$

Here,  $f(\mathbf{v})$  represents probability of finding a molecule of velocity  $\mathbf{v}$ ,  $w = (2k_B T/m)^{\frac{1}{2}}$ ,  $k_B$ : Boltzmann's constant ( $1.38 \times 10^{-23}$  J/K),  $T$ : gas temperature in Kelvin and  $m$  is molecular mass.



**Fig. 3.** Maxwellian velocity distribution with a bulk motion  $\mathbf{v}_0$ . The FWHM is proportional to  $\sqrt{T}$ .

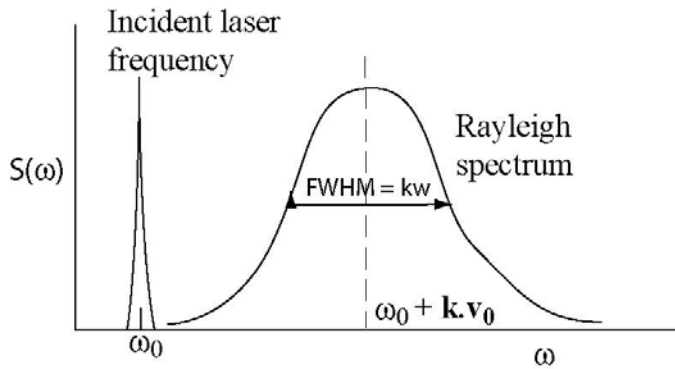
The Spectrum of the Rayleigh scattered light is:

$$S(\omega) = \int_{-\infty}^{\infty} d\tau \exp[i(\omega - \omega_0)\tau] \int_{-\infty}^{\infty} d^3v f(v) \exp[-i\mathbf{k} \cdot \mathbf{v} \tau] \quad (17)$$

$$= \frac{2\sqrt{\pi}}{wk} \exp\left[-\frac{(\omega - \omega_0 - \mathbf{v}_0 \cdot \mathbf{k})^2}{w^2 k^2}\right]$$

As expected, the integration involves 2 steps, once on velocity  $\mathbf{v}$  and second on time  $\tau$ . Figure 4 shows the spectral shape. The peak of the Rayleigh spectrum is shifted from that of the incident light by  $\mathbf{k} \cdot \mathbf{v}_0$ . Therefore a measure of this shift provides a measure of the bulk motion of the gas. Note that a component of velocity along the scattering vector  $\mathbf{k}$  is measured. The Full Width at Half Maxima (FWHM) is proportional to square root of gas temperature  $T$

$$FWHM = \frac{8\pi}{\lambda} \sqrt{\ln 2} \sqrt{\frac{2k_B T}{m}} \sin \frac{\theta}{2} \quad (18)$$



**Fig. 4.** Rayleigh scattered spectrum from Maxwellian velocity distribution.

Equations 11 and 18 provide the basis for aerodynamic measurement by Rayleigh scattering technique. In a nutshell, the total power of the scattered light is proportional to the gas density, the shift in the Rayleigh spectrum from that of the incident light is related to the bulk velocity and the half width of the spectrum is proportional to  $\sqrt{T}$ . The effect of neglecting Brillouin scattering is in the approximate description of temperature

dependence. Brillouin scattering (and the rotational Raman part) further broadens the spectra, yet density and bulk velocity measurements remain unaffected.

## Reference

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